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The Density, Magnetic Properties, Young's Modulus, and ΔE-Effect, and Their Change Due to Quenching in Ferromagnetic Iron-Aluminium Alloys. II

Young's Modulus and the ΔE-Effect*

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Synopsis

The ΔE-effect and Young's modulus in annealed and in quenched states of iron-aluminium alloys containing less than 17 percent aluminium have been measured with the method of magnetostrictive vibration. It is shown that the ΔE-effect increases to a marked extent while Young's modulus decreases by 2～3 percent with the formation of superlattice Fe_3Al and that low values of Young's modulus for alloys near Fe_3Al have no direct correlation with the existence of superlattice Fe_3Al.

I. Introduction

Iron-rich iron-aluminium alloys have been the subject of many investigations as typical ferromagnetic superlattice alloys. Young's moduli of these alloys have been measured with the statical bending method by Nishiyama(1) and by Masumoto and Saito(2), with the magnetostrictive vibration method by Kubo(3) and by T. Yamamoto(4), and with the statical interferometric method by Fukuroi and Shibuya(5). But, the ΔE-effect has not yet been observed.

Now, Young's modulus in unmagnetized state, E_0, of a ferromagnetic substance is different from that of a non-ferromagnetic one in that it involves a contribution from the displacements of domain walls and the rotation of magnetization vectors in domains in addition to the contribution from the cohesion, and hence it is structure-sensitive similarly to the initial magnetic susceptibility. Previous investigations(3,2,4,5) indicated a conspicuous minimum of E_0 near the Fe_3Al composition which has naturally been regarded as connecting to the existence of the Fe_3Al superlattice. But, whether this minimum of E_0 corresponds or not to a change in cohesional Young's modulus accompanied by the order-disorder transition in a

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(1) Z. Nishiyama, Sci. Rep. Tôhoku Univ., **18** (1929), 359.

(2) H. Masumoto and H. Saito, Nippon Kinzoku Gakkai-shi, **8** (1944), 359; Sci. Rep. RITU, **A3** (1951), 523.

(3) T. Kubo, Nippon Sugaku Buturi Gakkai-shi, **16** (1942), 426; **17** (1943), 449; Tôshiba Kenkyû Jihô, **18** (1943), 309 (both in Japanese).

(4) T. Yamamoto, Denki Tsûshin Gakkai-shi, **229** (1942), 273 (in Japanese).

(5) T. Fukuroi and Y. Shibuya, Sci. Rep. RITU, **A5** (1953), 405.

non-ferromagnetic substance cannot be decided from the measurements of E_0 alone. Thus, we have measured, with the method of magnetostrictive vibration, the ΔE-effect and Young's modulus and their change due to quenching in ferrromagnetic iron-aluminium alloys containing less than 17 percent aluminium. The study on the density and magnetic properties and their change due to quenching in these alloys made in connection with the present investigation was published previously(6).

The method, apparatus, and procedure of measurement were described previously(7,8,9). Specimens and heat-treatment(10) employed are the same as in the density and magnetic measurements(6) (cf. Table 1).

Table 1. Composition, saturation value of ΔE-effect, $(\Delta E/E_0)_s$, absolute value of negative ΔE-effect, $|(\Delta E/E_0)_-|$, and Young's modulus in unmagnetized state, E_0, for specimens of iron-aluminium alloys containing less than 17%Al in annealed and in quenched states.

No.	Specimen Mark	Composition wt.-%Al	Annealed state			Quenched state		
			$(\Delta E/E_0)_s \times 10^2$	$\|(\Delta E/E_0)_-\| \times 10^2$	$E_0 \times 10^{-12}$ dynes/cm^2	$(\Delta E/E_0)_s \times 10^2$	$\|(\Delta E/E_0)_-\| \times 10^2$	$E_0 \times 10^{-12}$ dynes/cm^2
1	1b	0.02	0.450	0.017	2.152	—	—	—
2	3a	1.22	0.525	0.051	2.063	—	—	—
3	4	1.83	0.592	0.050	2.040	—	—	—
4	5a	2.75	0.840	0.020	1.998	—	—	—
5	6a	4.05	1.185	0.104	1.933	—	—	—
6	10	7.94	4.00	0.68	1.736	—	—	—
7	11	9.53	2.85	0.87	1.678	—	—	—
8	12	11.16	7.60	0.27	1.549	2.412	0.52	1.656
9	13	12.07	13.14	0.43	1.684	2.140	0.14	1.898
10	14b	13.15	8.12	0.20	1.173	1.210	0.46	1.294
11	15	13.92	5.46	0.30	1.486	1.110	0.06	1.574
12	16a	14.15	8.00	0.27	1.452	1.247	0.01	1.546
13	17	14.70	6.05	0.38	1.459	1.005	0.14	1.560
14	18	16.00	2.385	0.060	1.567	0.271	0.06	1.615
15	19	16.96	0.500	0.025	1.536	0.216	—	1.572

II. The ΔE-Effect

The relations between the change of Young's modulus with magnetization, relative to that in unmagnetized state, $\Delta E/E_0$, and the (effective) magnetic field, H, are shown in Figs. 1(a) ~(c). The low-field portions of them are shown separately in Figs. 2(a)~(c).

Alloys containing less than about 4 percent aluminium show the secondary

(6) M. Yamamoto and S. Taniguchi, Nippon Kinzoku Gakkai-shi, **17** (1953), 529 and 532; Sci. Rep. RITU, **A 8** (1956), 112.
(7) M. Yamamoto, Sci. Rep. Tôhoku Univ., **27** (1938), 115; Nippon Kinzoku Gakki-shi, **2** (1938), 495.
(8) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **5** (1941), 167; Sci. Rep. Tôhoku Univ., **13** (1943), 101.
(9) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **6** (1942), 331; Sci. Rep. RITU, **A3** (1951), 308.
(10) The annealing adopted (heated for 2 hours at 1000°C and then furnace-cooled in a vacuum) may not be perfect, but Young's modulus seems not to change appreciably by cooling more slowly than ours, since the rate of formation of superlattice Fe_3Al is fairly rapid(3).

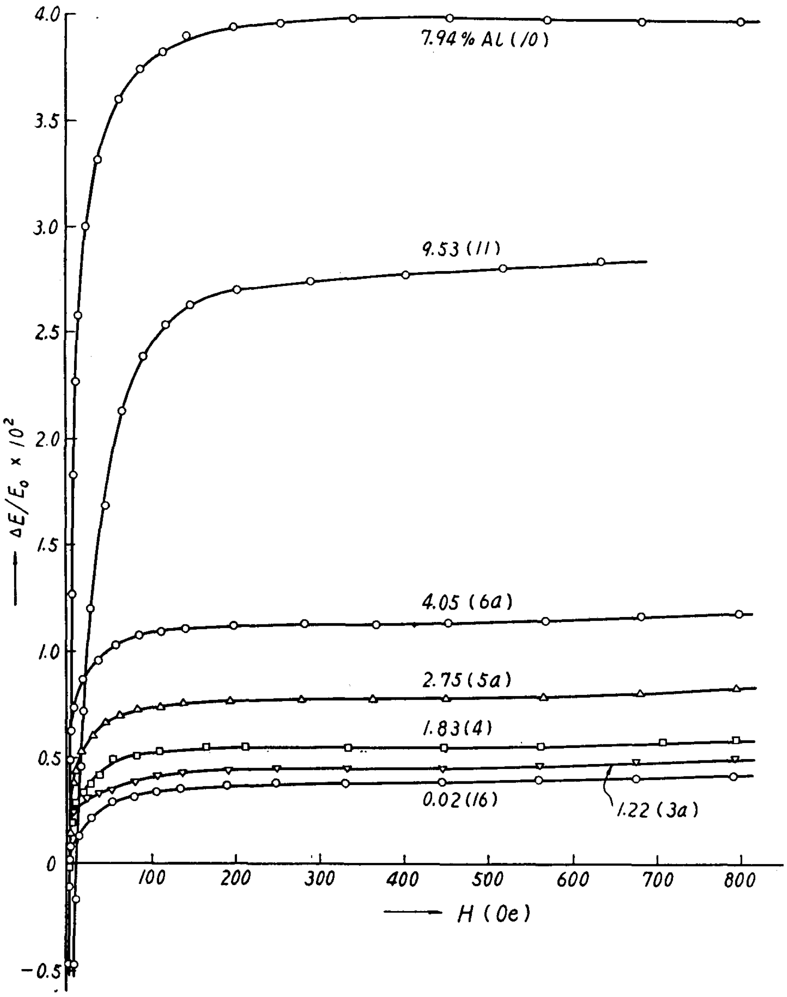

Fig. 1(a). $\Delta E/E_0$–H curves of *annealed* iron-aluminium alloys containing less than 10 percent aluminium.

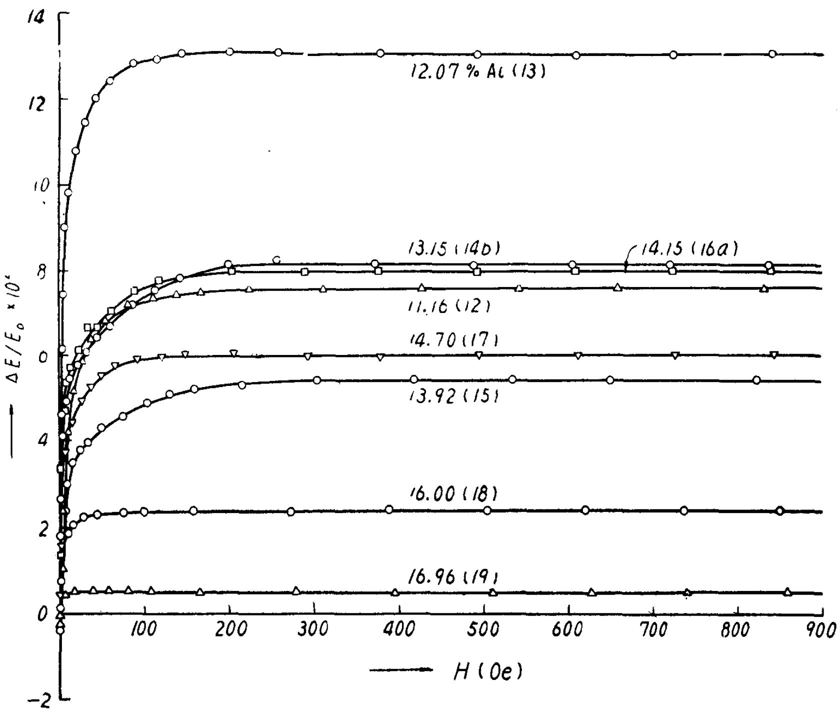

Fig. 1(b). $\Delta E/E_0$–H curves of *annealed* iron-aluminium alloys containing 11 to 17 percent aluminium.

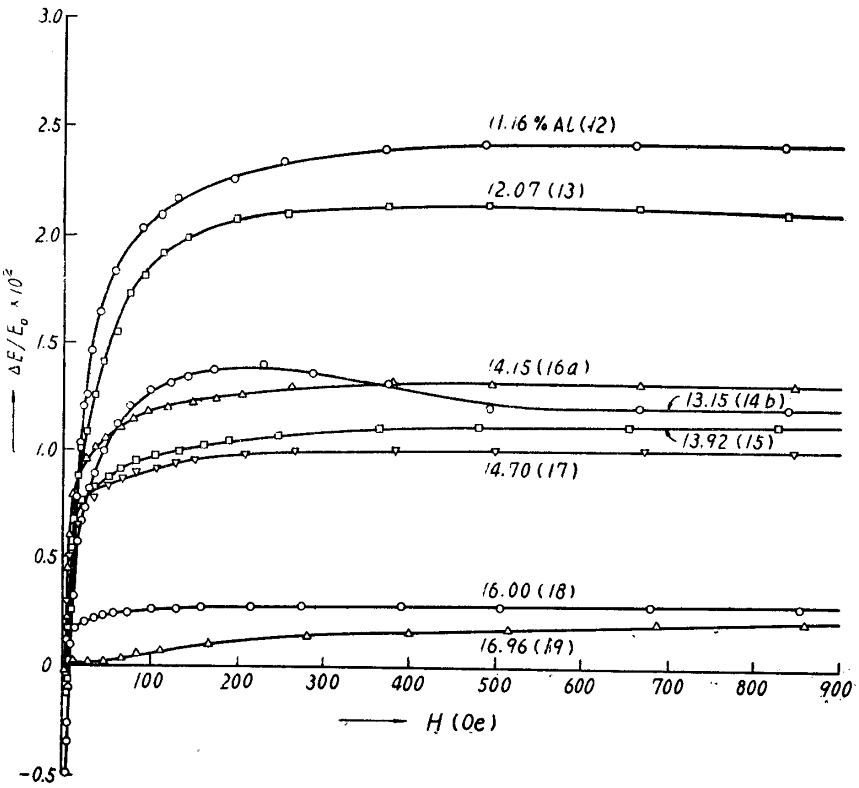

Fig. 1(c). $\Delta E/E_0$-H curves of *quenched* iron-aluminium alloys containing 11 to 17 percent aluminium.

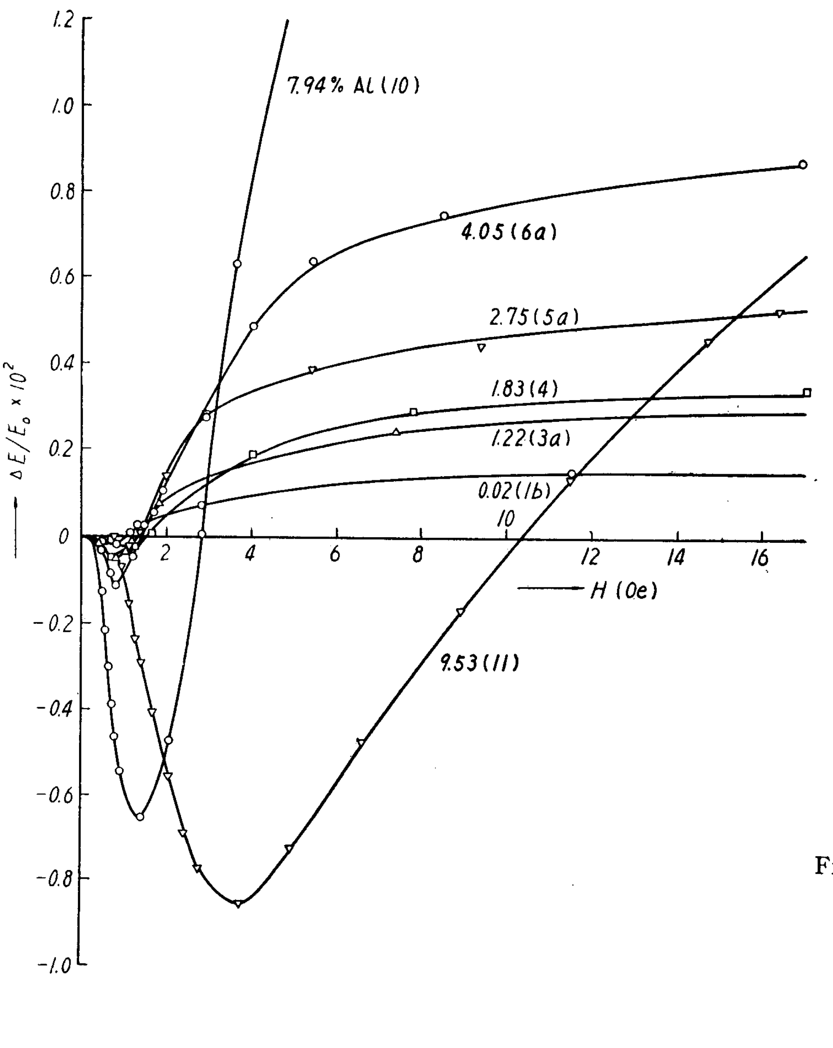

Fig. 2(a). $\Delta E/E_0$–H curves at low fields of *annealed* iron-aluminium; alloys containing less than 10 percent aluminium.

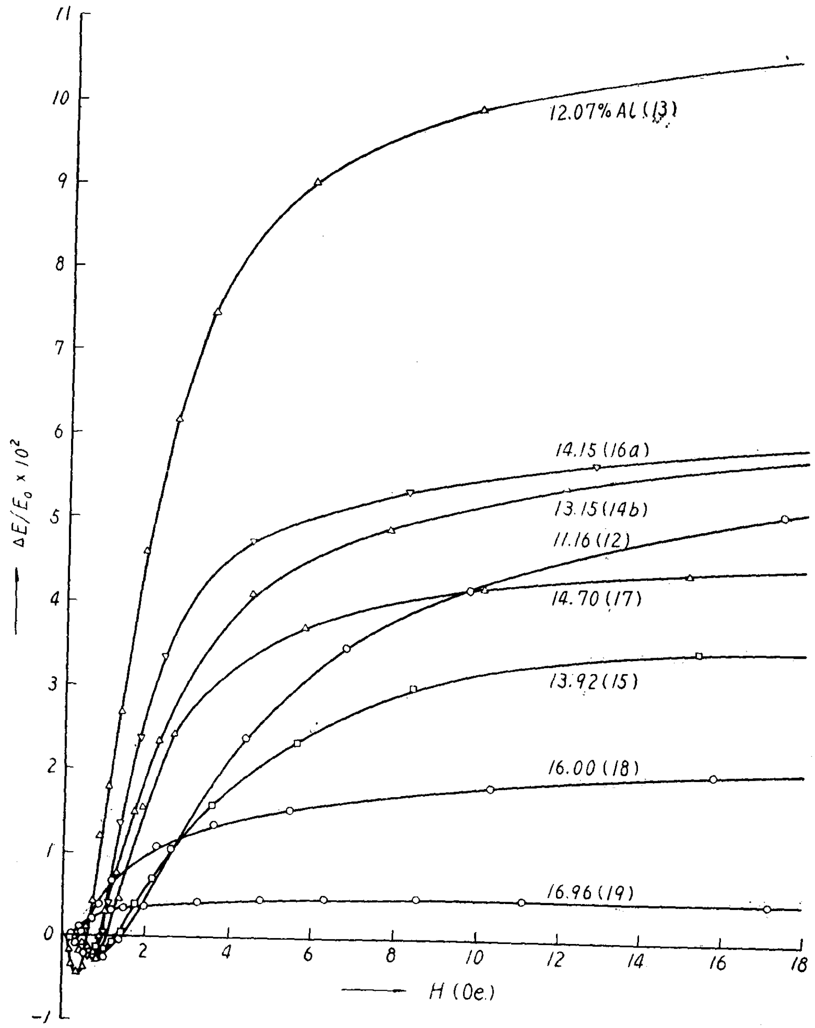

Fig. 2(b). $\Delta E/E_0$–H curves at low fields of *annealed* iron-aluminium alloys containing 11 to 17 per-cent aluminium.

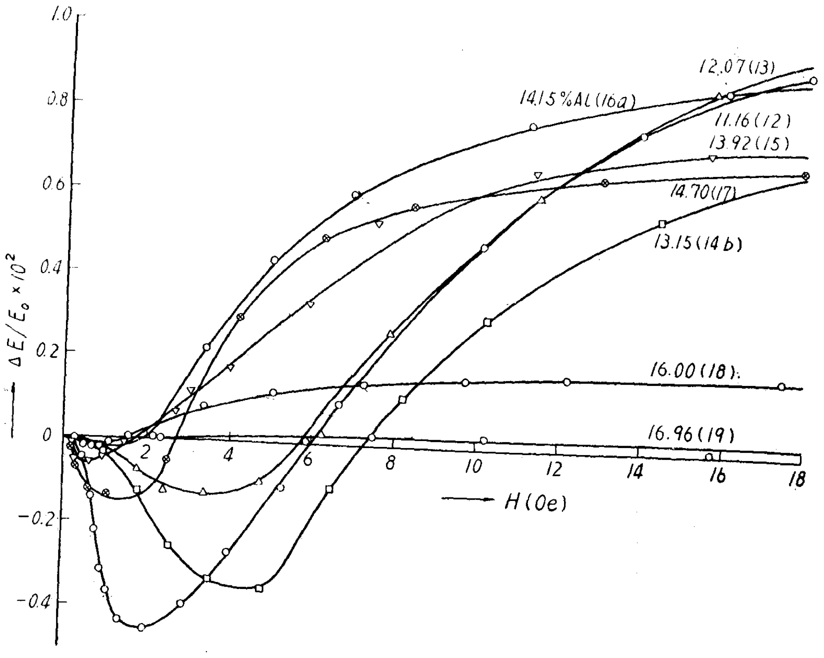

Fig. 2(c). $\Delta E/E_0$–H curves at low fields of *quenched* iron-aluminium alloys containing 11 to 17 per-cent aluminium.

increase of the ΔE-effect at high fields (see Fig. 1(a)). This phenomenon was previously found by one of the authors (Yamamoto) with iron[7,8,11,12], nickel[8,11,13,14], nickel-copper[11], iron-cobalt[12], iron-nickel[13], and nickel-cobalt[14] alloys, and considered as associated with the ferromagnetic anisotropy energy. Moreover, in the field range of saturation, Young's modulus in annealed state of 16.96%Al alloy shows a slight decrease (Fig. 1(b)) and that in quenched state of 13.15%Al alloy exhibits a considerable decrease (Fig. 1(c)). Similar effect was already observed by Yamamoto[13] with some iron-nickel alloys. This effect may be interpretable as associated with the volume magnetostriction[15].

Furthermore, the ΔE-effect in ferromagnetic iron-aluminium alloys, irrespective of heat treatment, shows a small negative value at low fields and soon changes to a positive value, as seen from Fig. 2(a)~(c). The composition dependences of the (negative) minimum value of the ΔE-ffect, $(\Delta E/E_0)_-$, and of the corresponding field, H_-, are shown in Fig. 3. Such a negative ΔE-effect at low fields was frequently observed with iron[12], nickel-copper[11], iron-cobalt[12], iron-nickel[13,16], nickel-cobalt[14], and iron-nickel-cobalt alloys[17]. It may be seen from the contrast with

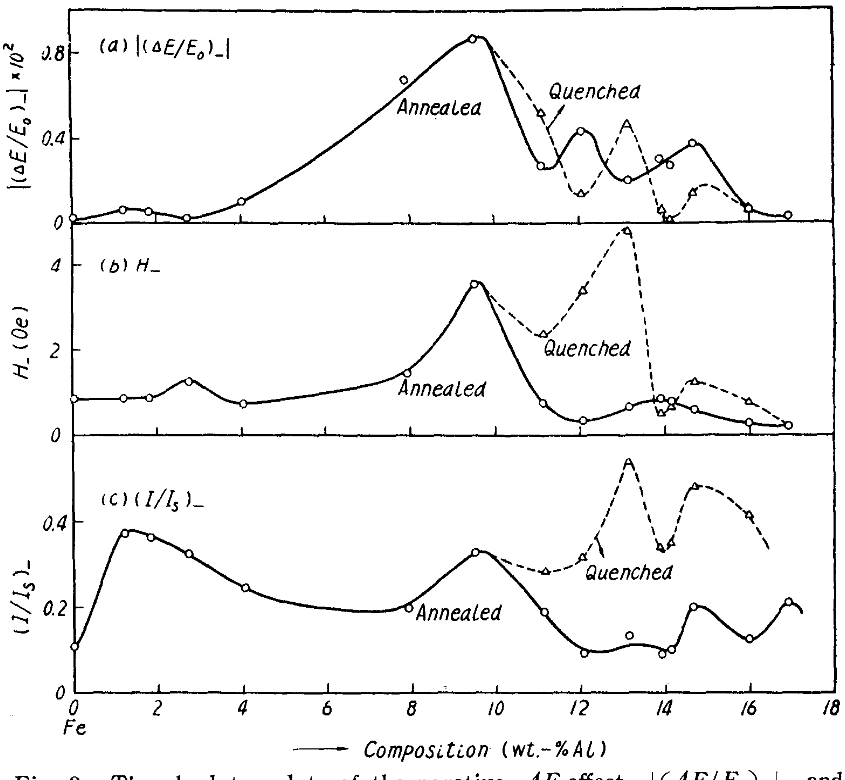

Fig. 3. The absolute valute of the negative ΔE-effect, $|(\Delta E/E_0)_-|$, and the field and reduced magnetization at which $(\Delta E/E_0)_-$ is attained, H_- and $(I/I_s)_-$,* as functions of the composition in iron-aluminium alloys in annealed and in quenched states.

(11) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **6** (1942), 249 ; Sci. Rep. RITU, **A6** (1954), 446.
(12) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **6** (1942), 581 (in Japanese).
(13) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **7** (1943), 467 (in Japanese).
(14) M. Yamamoto, Nippon Kinzoku Gakkai-shi, **12** (1948), No. 2–3. M. Yamamoto and S. Taniguchi, ibid., **B15** (1951), 337 ; Sci. Rep. RITU, **A6** (1954), 35.
(15) R. Kimura and K. Kondô, Tokyô Daigaku Rikôgaku-Kenkyûsho Hôkoku, **1** (1947), 69 (in Japanese).

the magnetization curves shown in the preceding paper[6] that H_{-} corresponds roughly with the inflexion point of a magnetization curve, as pointed out previously by Yamamoto[9,12~15].

Figs. 4(a)~(c) show relations between the ΔE-effect and reduced magnetization, namely the ratio of the intensity of magnetization to the saturation magnetization, I/I_s. The composition dependence of the reduced magnetization corresponding to $(\Delta E/E_0)_{-}$, $(I/I_s)_{-}$, is as shown in Fig. 3(c).

As seen from the $\Delta E/E_0$-H curves shown in Fig. 1(a), at the maximum measuring field (about 800 oersteds), the ΔE-effects of low-aluminium alloys do not attain to saturation completely but are quite near to it, so that the values of ΔE-effect for the maximum measuring field may be regarded as the saturation ΔE-effect, $(\Delta E/E_0)_s$. $(\Delta E/E_0)_s$ as a function of the composition is shown by solid

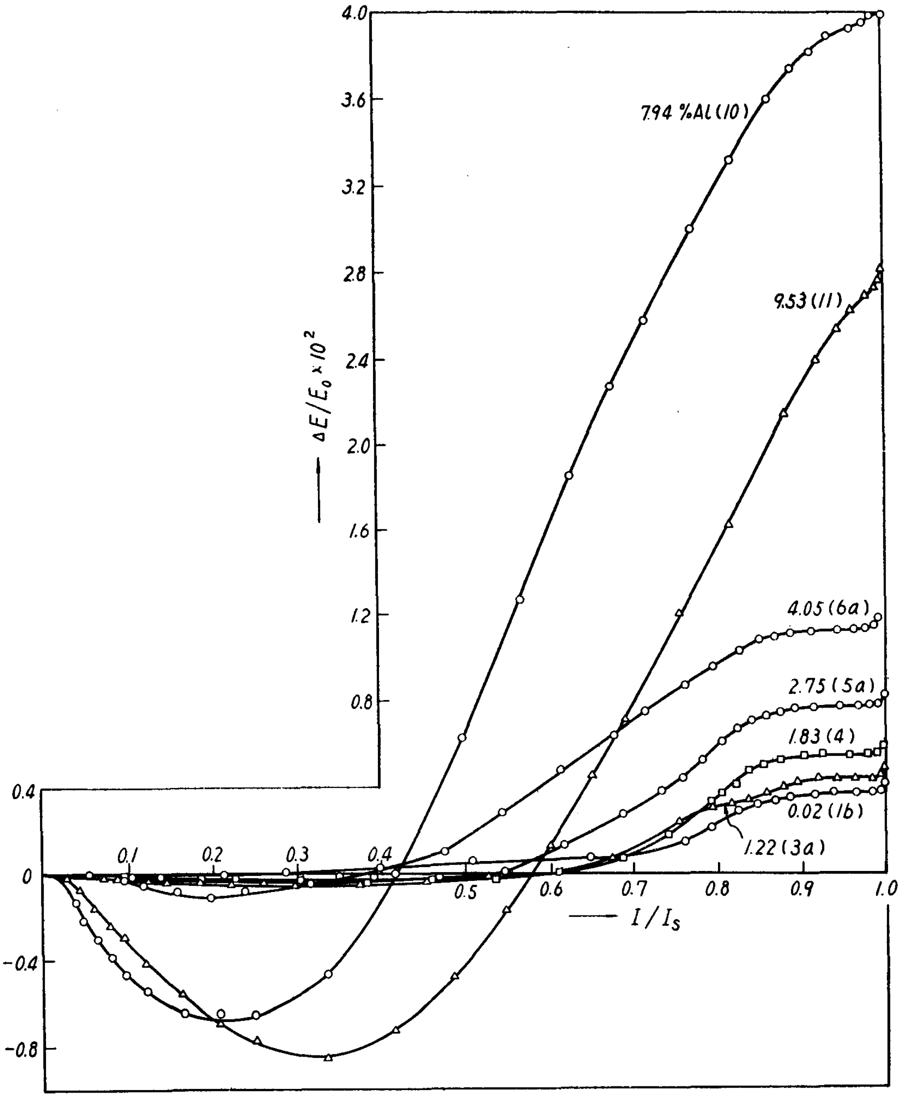

Fig. 4(a). $\Delta E/E_0$-I/I_s curves of *annealed* iron-aluminium alloys containing less than 10 percent aluminium.

(16) H. J. Williams, R. M. Bozorth and H. Christensen, Phys. Rev., **59** (1941), 1005.

(17) N. Kunitomi, J. Phys. Soc. Japan, **8** (1953), 26.

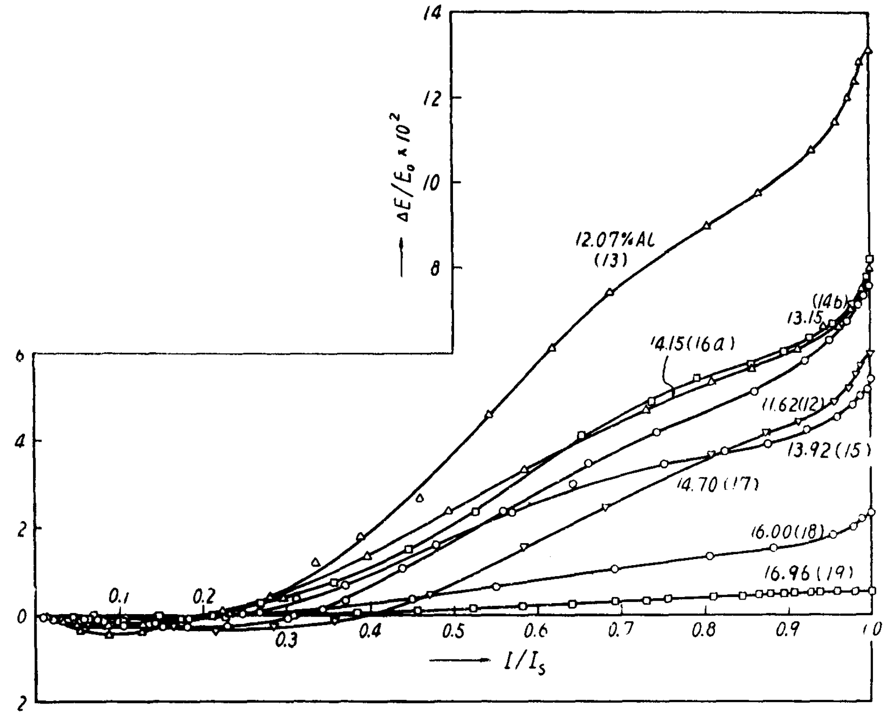

Fig. 4(b). $\Delta E/E_0$–I/I_s curves of *annealed* iron-aluminium alloys containing 11 to 17 percent aluminium.

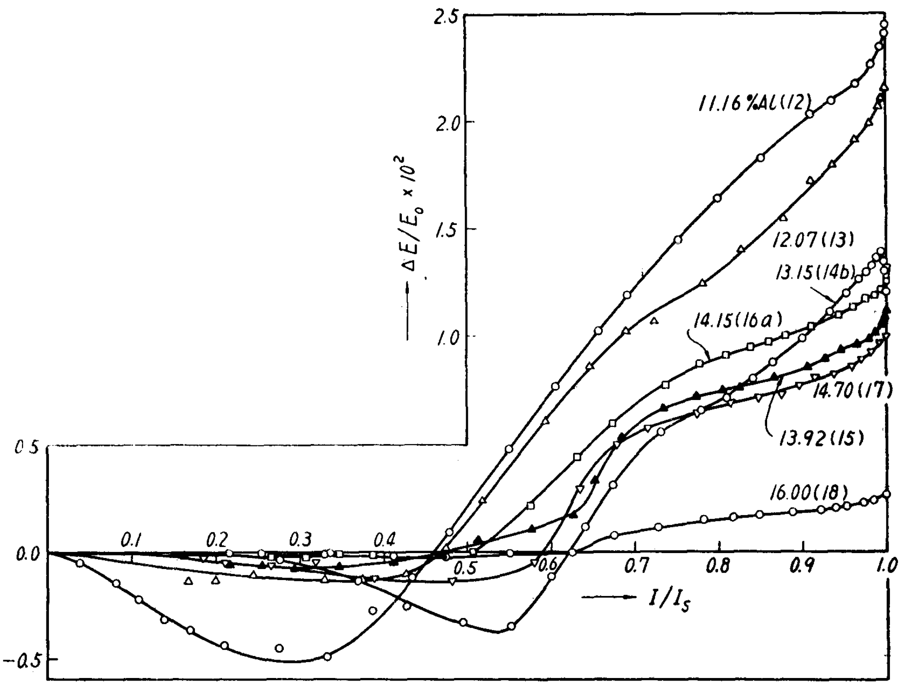

Fig. 4(c). $\Delta E/E_0$–I/I_s curves of *quenched* iron-aluminium alloys containing 11 to 17 percent aluminium.

lines in Fig. 5. $(\Delta E/E_0)_s$ for annealed state exhibits three maxima of about 4, 13 and 8 percent, respectively, at about 8, 12 and 14 percent aluminium and then vanishes at about 17.5 percent aluminium. Quenching from 700°C decreases considerably the saturation ΔE-effect of high-aluminium alloys, obliterating the two

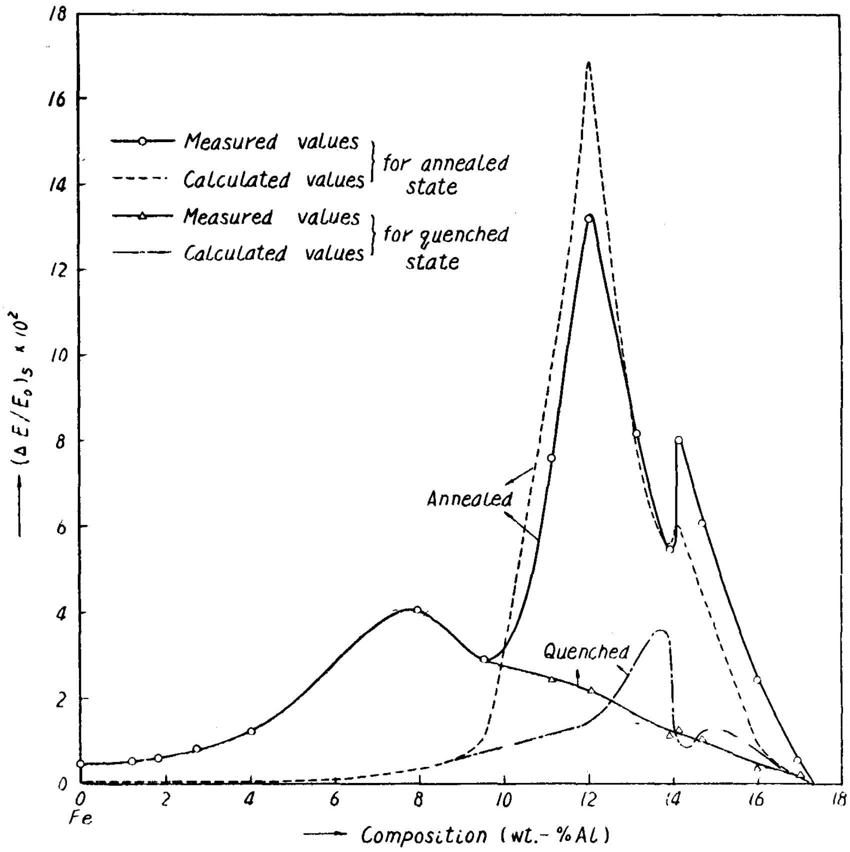

Fig. 5. $(\Delta E/E_0)_s$ as a function of the composition in iron-aluminium alloys in annealed and in quenched states.

maxima at about 12 and 14 percent aluminium.

Fig. 5 contains, further, the values of $(\Delta E/E_0)_s$ calculated from a semi-theoretical and semi-experimental formula, which has been found to agree fairly with experiments for most cases[13~15]:

$$(\Delta E/E_0)_s = 0.7\chi_0\lambda_s^2E_0/I_s^2 \qquad (1)$$

where χ_0 is the initial susceptibility, I_s the saturation magnetization and λ_s the saturation magnetostriction. For calculation we used our measured values for χ_0[6], I_s[6], and E_0 (cf. section III) and values interpolated from the measured values of Masumoto and Saito[18] for λ_s. For alloys containing more than about 10 percent aluminium, the calculated and measured values are in good agreement with each other especially for annealed state, and also in fairly good agreement for quenched state, which indicates that the large ΔE-effect of annealed alloys containing more than about 11 percent aluminium are mainly caused by the large magnetostriction of them[18].

It is to be noted, however, that a large difference exists between the calculated and measured values for low aluminium alloys containing some 8 percent aluminium. Theoretically, Eq. (1) is an approximate formula obtained by averaging

(18) H. Saito, Nippon Kinzoku Gakkai-shi, **B14** (1950), No. 5; H. Masumoto and H. Saito, Sci. Rep. RITU, **A4** (1952), 338.

with Reuss' method only contributions from the displacements of non-180° domain walls in the case where the magnetocrystalline anisotropy energy predominates over the magnetic strain energy(11). Accordingly, λ_s in Eq. (1) is really the saturation magnetostriction in the direction of easy magnetization, λ_e, and λ_e is equal to λ_s only if the magnetostriction is isotropic. In fact, Eq. (1) has been found to coincide fairly well with measurement for cases where the magnetostriction is approximately isotropic, namely for nickel(11), nickel-copper(11,19), iron-cobalt(12,20), nickel-iron(13) and low-cobalt nickel-cobalt(14) alloys. Thus, the above-mentioned inconsistency between the measured and calculated values of $(\Delta E/E_0)_s$ for alloys containing less than about 10 percent aluminium may be considered as due to the high anisotropy in magnetostriction for these alloys, which may be inferred from their magnetostriction curves(21).

III. Young's modulus

Young's modului in unmagnetized and in magnetically saturated states, E_0 and E_s, as functions of the composition are shown in Fig. 6. With increasing aluminium content, E_s decreases at first linearly and, as the aluminium content exceeds over about 11 percent, it falls rapidly and makes a conspicuous minimum at about

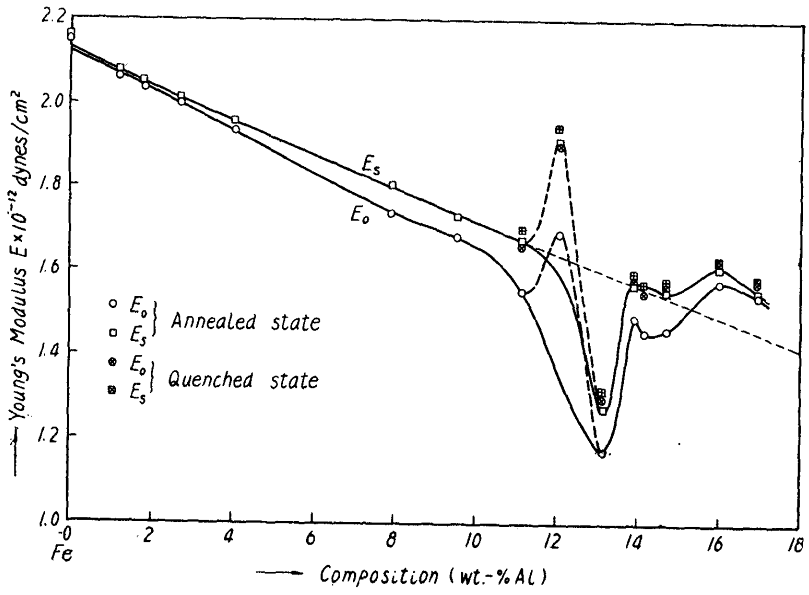

Fig. 6. Young's moduli at unmagnetized and at magnetically saturated states, E_0 and E_s, as functions of the composition in iron-aluminium alloys in annealed and in quenched states.

(19) The approximate isotropy of the magnetostriction of nickel-copper alloys may be quite sure, since the magnetostriction is nearly isotropic even for nickel and with increasing copper content the magnetostriction curves of polycrystalline alloy specimens become less and less steep, keeping the similar form and negative λ_s with decreasing absolute value.

(20) K. Azumi, Un-yu Gijutsu-Kenkyûsho Hôkoku, **4** (1954), 1 has shown that the magnetostriction of body-centered cubic iron-cobalt alloys containing more than about 40 percent cobalt is approximately isotropic.

(21) K. Honda, H. Masumoto, Y. Shirakawa and T. Kobayashi, Nippon Kinzoku Gakkai-shi, **12** (1948), No. 7-12; Sci. Rep. RITU, **A1** (1949), 341.

13 percent aluminium, then recovering to the extension of the linear decrease in the low-aluminium range (thin dotted line ln Fig. 6) at about 14 percent aluminium. Afterwards, E_s increases slightly and again decreases. E_0 goes approximately parallel to E_s.

It is to be noted that values of E_0 and E_s for 12% Al alloy lie far above the just-mentioned curves. An E_0 vs. composition curve obtained by Kubo[3] seems to show a maximum near 12 percent aluminium (Fig. 7[22]). Further, a minimum of the E_0 vs. composition curve for annealed state near 13 percent aluminium was found first by Kubo[3] and since then it was confirmed frequently[2,4,5] (Fig. 7).

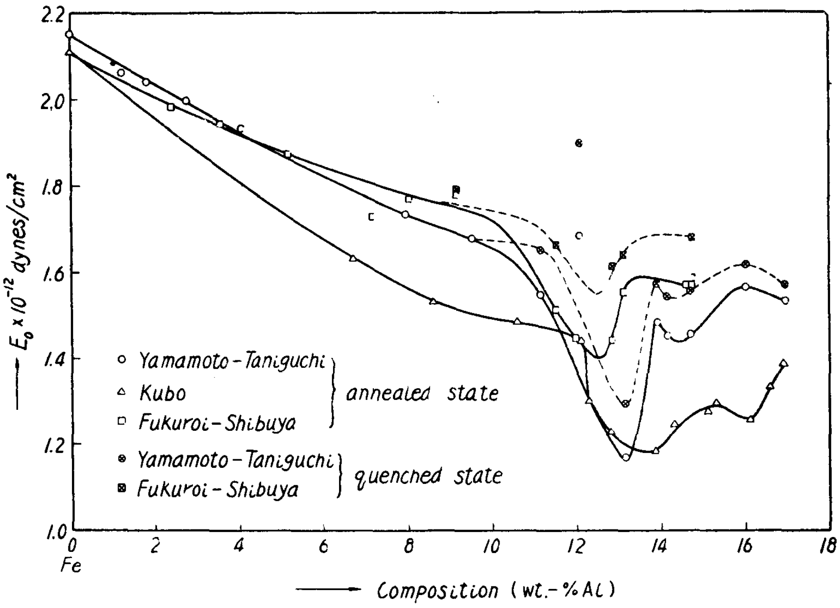

Fig. 7. Young's modulus at unmagnetized state, E_0, as dependent upon the composition in iron-aluminium alloys in annealed and in quenched states, compared with the data obtained by Kubo and by Fukuroi and Shibuya.

Quenching from 700° raises both E_0 and E_s. The increase in E_0 is conspicuous in agreement with an observation by Fukuroi and Shibuya[5], while the increase in E_s amount to only 2~3 percent. In other words, Young's modulus of Fe_3Al decreases with ordering in contradiction to cases of CuZn[23], Cu_3Au[24], Cu_3Pd[25], and Ni_3Fe[26].

The relation between the relative increase of E_s by quenching and composition is roughly inverse to that between the relative increase of density by quenching

(22) Kubo's data are low, since they were determined using Nishiyama's[1] X-ray data for the lattice constant.

(23) W. Köster, Z. Metallkde, **32** (1940), 145; J. S. Rinehart, Phys. Rev., **58** (1940), 365; W. A. Good, Phys. Rev., **60** (1941), 605.

(24) H. Röhl, Ann. Physik, **18** (1933), 155; W. Köster, Z. Metallkde, **32** (1940), 145; S. Siegel, Phys. Rev., **57** (1940), 537; J. Chem. Phys., **8** (1940), 860; N. W. Lord, J. Chem. Phys., **21** (1953), 692; S. Umekawa, Nippon Kinzoku Gakkai-shi, **18** (1954), 449.

(25) H. Röhl, Ann. Physik, **18** (1933), 155.

(26) M. Yamamoto, T. Suzuki and S. Taniguchi, to be published.

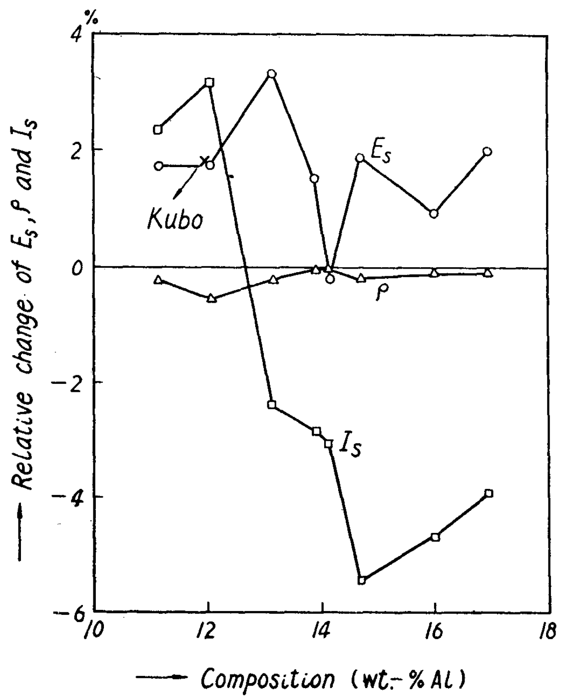

Fig. 8. Relative changes due to quenching from 700°C of Young's modulus at magnetically saturated state, E_s, the density, ρ, and the saturation magnetization, I_s, as functions of the composition in iron-aluminium alloys containing 11 to 17 percent aluminium.

and composition[6], as seen from Fig. 8. Our investigation[6] indicates that the composition where the relative change of the saturation magnetization by quenching changes its sign, namely about 12~13 percent aluminium, is the boundary between the disordered α and ordered FeAl phases and near this boundary E_0 and E_s undergo conspicuous changes as mentioned above. It is to be noted, further, that, as seen from Fig. 6, the minima at about 13 percent aluminium of E_0 and E_s are never obliterated by quenching, indicating that they are not directly connected to the existence of the superlattice Fe_3Al.

Summary

The ΔE-effect and Young's modulus in annealed and in quenched states of iron-aluminium alloys containing less than 17 percent aluminium have been measured with the method of magnetostrictive vibration. In the whole composition range a small negative ΔE-effect has been observed at low fields in annealed state as well as in quenched state, and, in addition, a secondary rise of the ΔE-effect has been found with alloys containing less than 4 percent aluminium. The saturation ΔE-effect in annealed state shows three maxima at about 8, 12 and 14 percent aluminium, of which the second and third are obliterated by quenching, indicating that the ΔE-effect increases to a greater extent with the formation of superlattice Fe_3Al corresponding to a large increase of magnetostriction with ordering. Young's moluli in annealed state as well as in quenched state exhibit commonly deep minima at about 13 percent aluminium, which are never obliterated by quenching, indicating that they have no direct correlation with the existence of superlattice Fe_3Al. Young's modulus in magnetically saturated state in alloys near Fe_3Al increases by 2 or 3 percent due to quenching. In other words, Young's modulus of Fe_3Al decreases with ordering, contrary to cases of CuZn, Cu_3Au, Cu_3Pd and Ni_3Fe.